Bilayer oscillation of subband effective masses in Pb/Ge(111) thin-film quantum wells

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Subband dispersions of quantum-well states in Pb thin films on Ge(111) have been measured with angle-resolved photoemission spectroscopy. The effective masses at the surface zone center exhibit a bilayer oscillation with thickness, in both magnitude and sign. This behavior is attributed to a strong interaction between Pb quantum-well subbands and the Ge valence maximum near the Fermi level, which occurs about every two monolayers. © *2010 American Institute of Physics*. [doi:10.1063/1.3358130]

The subband dispersions of quantum-well states (QWS) in thin films are crucial to the properties of these films, especially the electric conductance.[1] According to the Drude model, the conductivity is inversely proportional to the effective masses of electrons. Furthermore, in the two-dimensional (2D) Boltzmann regime,[1] two important elements for the conductivity—the Fermi velocity and the density of states at the Fermi level—are intimately related to the effective masses of the QWS subbands. Hybridization interaction between the film and the substrate has been found to affect the QWS subbands in most thin film systems on semiconductor substrates,[2,3] and the energy position of the semiconductor valence maximum plays an important role in the value of the effective mass, which becomes much enhanced when the QWS at the surface zone center approach the valence maximum of the substrate.

A thin film of Pb exhibits a bilayer oscillatory behavior in various properties,[4] because the QWS cross the Fermi level about every two monolayers, causing a bilayer modulation of both the surface energy and the density of states at the Fermi level. If the valence maximum of the semiconductor substrate is also near the Fermi level, the QWS are likely to interact with the valence maximum every two monolayers. As a result of this interaction, the effective masses of QWS subbands would exhibit corresponding bilayer oscillations with thickness. A Ge substrate is effective for this research as the valence maximum of Ge is near the Fermi level. We have investigated the effective masses of QWS subbands in thin films of Pb on highly doped *n*-type Ge(111) because the valence maximum becomes increasingly near the Fermi level with the doping effect.[5] The key of this work is to investigate how the effective masses of the QWS subband are modulated through the interaction with the substrate for a varied thickness.

Angle-resolved photoemission measurement was performed with an energy analyzer (Scienta R3000) and unpolarized He I light of photon energy 21.2 eV. Some additional data were recorded at beamline 21B1-U9 in the National Synchrotron Radiation Research Center in Taiwan; the results are consistent. A highly doped ($\sim 10^{18}$ cm^{-3}) *n*-type Ge(111) wafer served as sample. The procedure to prepare a clean Ge(111)-c(2×8) surface is described elsewhere.[6] An overlayer uniform Pb film was formed on depositing Pb onto a Pb/Ge(111)-$\sqrt{3}\times\sqrt{3}$ R30° structure kept at −150 °C. The film thickness was calibrated through a standard procedure.[7]

Figure 1 shows the energy distribution curves (EDC) of QWS at normal emission for films of thickness from 0 to 11 ML Pb films on the Pb/Ge(111)- $\sqrt{3}\times\sqrt{3}$ R30° surface. Each of EDC was measured after deposition of Pb for one minute at a steady rate ~0.2 ML/min. The QWS peaks have maximum intensity at integer layers, of which the corresponding EDC are indicated with blue color. It is clear from the spec-

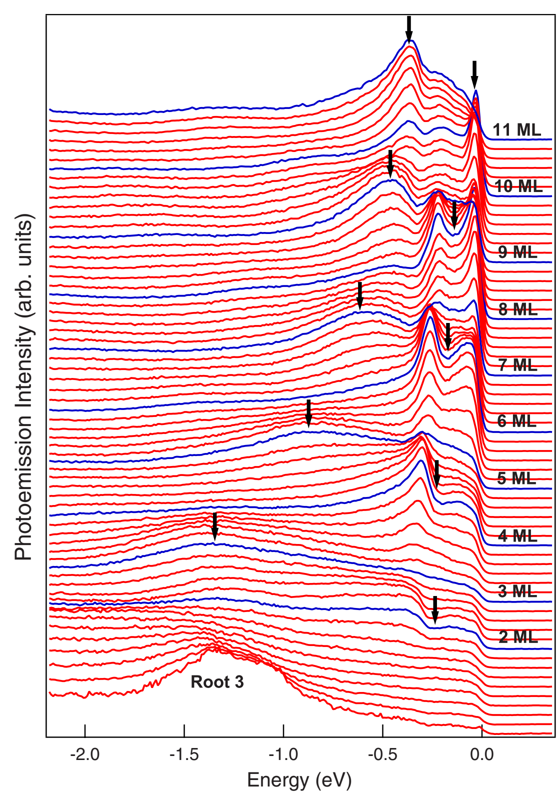

FIG. 1. (Color online) Photoemission spectra at normal emission taken from Pb films of thicknesses from 0 to 11 ML on the Pb/Ge(111)-$\sqrt{3}\times\sqrt{3}$ R30° surface.

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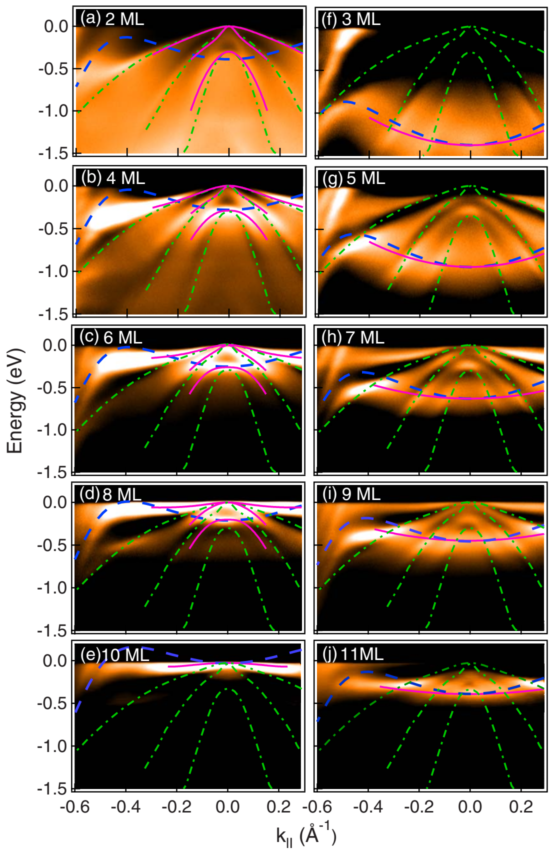

FIG. 2. (Color online) Angle-resolved photoemission spectra for Pb films ranging from 2 to 11 ML in the symmetry direction from $\bar{\Gamma}$ to $\bar{K}$ are presented as a gray-scale image. The vertical axis represents energy, and the horizontal axis represents the in-plane momentum $k_{\parallel}$.

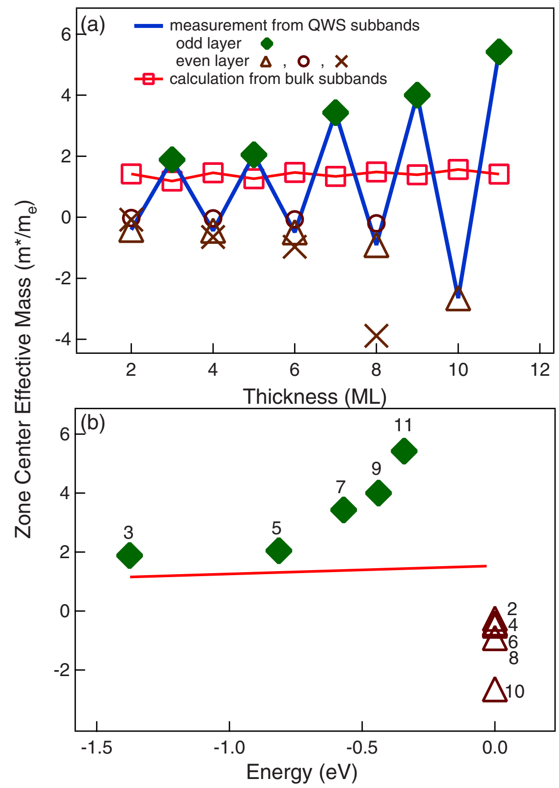

FIG. 3. (Color online) (a) Thickness dependence and (b) energy dependence of effective masses of QWS subbands for Pb films ranging from 2 to 11 ML. The numbers indicate the corresponding thickness.

trum that, when the intensity of the QWS peak corresponding to N ML decreases, the peak corresponding to $N+1$ ML begins to emerge. This behavior demonstrates that the films grow layer by layer. According to our thickness calibration, the first QWS peak to emerge is at 2 ML, indicating a critical thickness. Moreover, the QWS associated with even layers are nearer the Fermi level than those associated with odd layers. At even layers, the QWS peaks split into two components because of a strong interaction with Ge band edges at the valence maximum near the Fermi level. This strong interaction further affects the in-plane dispersion of QWS, as revealed by the effective mass. The origin of the interaction is hybridization.[6] Because the Ge valence maximum is near the Fermi level, the QWS observed are resonances. The QWS peaks indicated with the arrows in Fig. 1 are those for which we have investigated the subband dispersions.

Figure 2 shows the 2D angle-resolved photoemission spectra of QWS subbands in the symmetry direction from $\bar{\Gamma}$ to $\bar{K}$ for Pb films of thickness 2–11 ML. The subband dispersions of the odd and even layers about the surface zone center ($k_{\parallel}=0$) differ substantially. Near the zone center, the band dispersions of the even layers are downward parabola-like curves, whereas, for odd layers, the Pb band dispersions are similar to upward parabolas with distortion near the Ge band edges at the off-normal positions. To elucidate the interaction between the Ge valence maximum and the Pb QWS bands, the calculated Ge hole-band edges, heavy hole (HH), light hole (LH), and split-off (SO) band edges (green dashed-dotted curves),[8] and the expected subbands of QWS (blue dashed curves) for free-standing Pb films, based on a tight-binding calculation of the bulk band structure of Pb,[9] are superimposed on the data. Through fitting the EDC or momentum dispersion curves for QWS, we extracted the QWS subbands. They were then further fitted with the model (pink solid curves)

$$\mathcal{E}(k_{\parallel}) = \mathcal{E}(0) + \frac{\hbar^2 k_{\parallel}^2}{2m^*}\frac{1+ak_{\parallel}^2}{1+bk_{\parallel}^2}, \qquad (1)$$

in which a Padé function (ratio of polynomials) is employed to account for the band distortion; a, b, and the zone-center effective mass m^* are treated as fitting parameters. The resulting effective masses as a function of thickness are shown in Fig. 3(a), which exhibits a bilayer oscillation with negative values at even layers and positive values at odd layers. The effective masses derived from the calculated bulk bands, represented with red squares, with the same energies at the zone center as those of the measured QWS subbands, also show a bilayer oscillation but only in magnitude and with a 180° phase reversal. For each QWS subband, the absolute values of the zone center effective mass, m^*, increase substantially with increasing thickness of the film. This result is attributed to the degree of hybridization with the substrate bulk band edges. We consider two scenarios for the dependence of the positive and negative effective masses on thickness. For odd layers, as the film thickness increases, the QWS shift toward the Ge valence-band maximum. The nearer that the QWS moves to the Ge valence-band maximum, the flatter is the subband dispersion of the QWS, as pulled by the Ge band edges with negative curvatures. The effective mass of QWS thus becomes much larger than the corresponding value of the bulk subbands near the energy position of the Ge valence-band maximum, as shown by the filled rhombuses in Fig. 3(b). For the even layers, the energy positions of QWS are near the Ge valence-band maximum, and the shift of energy of the QWS with thickness is negligible. At thickness 2 ML, as seen in Fig. 2(a), three QWS subbands match the calculated HH, LH, and SO band edges satisfactorily. The density of states of QWS at 2 ML is hence dominated by those of the Ge bulk band edges.[6] With increasing Pb thickness, the Ge bulk band edges become less dominant; the three subband dispersions of QWS hence turn to expand from negative curvatures, leading to the increasing absolute values of the negative masses, as shown by hollow triangles, circles, and crosses in Fig. 3.[10] In Fig. 3(b), the red curve represents the result calculated from the projected bulk band structure of Pb, showing a smooth and negligible variation through the Ge valence maximum. Behavior of this kind is found in a system such as Pb/highly oriented pyrolytic graphite (HOPG),[11] in which the substrate has a large energy gap near the zone center so that the valence band edge of the substrate has almost no effect on the QWS subbands. In contrast, for the system Pb/Ge(111), the Ge valence maximum has a strong interaction with the Pb QWS bands. Because of the bilayer Fermi-level crossing of the QWS, and the proximate position of the Ge valence maximum to the Fermi level, the subband effective masses of QWS are modulated every two layers by the negative curvature of the Ge valence maximum.

In conclusion, we have investigated the subband effective masses of QWS at the surface zone center for Pb films on Ge(111) at thicknesses from 2 to 11 ML. The subband effective masses at the zone center are positive at odd layers but negative at even layers. According to Fig. 2, the QWS subbands that we investigated at even layers mostly form hole packets around the surface zone center. These holelike bands are similar to the valence-hole bands of semiconductors in the sense that the charge of the band responds to an applied electric or magnetic field in a manner opposite to that of the electronlike band. Therefore, the sign of the Hall coefficient might alter in an interesting bilayer manner for Pb films on Ge(111). Even though, for the transport properties of thin films, the entire QWS subbands that cross the Fermi level over all the surface Brillouin zone must be taken into account rather than the local area around the surface zone center,[1] our results imply the possibility of manipulating the thickness-dependent conductance of the thin films through a subtle match between the thin film and the substrate.

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[10] For even layers, hollow triangles, circles, and crosses represent the three QWS subbands corresponding to the HH-, LH-, and SO-like bands at 2 ML. In Fig. 3(a), the effective masses of the LH- and SO-like bands are not included at 10 ML because of their small intensity. In Fig. 3(b), for clarity, only the effective masses of HH-like band are included.
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